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Models for Calculating the Effects of Isotopic Exchange, Radioactive Decay, and of Recycle in Removing Iodine from Gas and Liquid Streams

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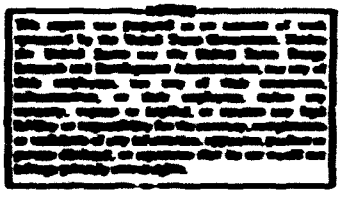

MODELS FOR CALCULATING THE EFFECTS OF ISOTOPIC EXCHANGE, RADIOACTIVE DECAY, AND OF RECYCLE IN REMOVING IODINE FROM GAS AND LIQUID STREAMS

Wallace Davis, Jr.

SEPTEMBER 1975

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ABSTRACT

Different decontamination factors for ^{129}I and ^{131}I are frequently invoked in environmental impact reports concerned with nuclear fuel recycle. Selected differences, or ratios, have not been justified on the basis of mathematical models or experimental data. This report presents a description of the origins of these differences in terms of isotopic exchange and material balance equations for the short- and long-lived (or stable) isotopes. The ratios of decontamination factors can be calculated when there is complete attainment of isotopic exchange between gas- or liquid-phase iodine and iodine sorbed by a solid or liquid. If there is no exchange, decontamination factors are isotope-independent unless material recycle occurs within the system. Between these extremes, there can be decontamination factors whose explanation requires experimental determination of the extent of exchange. The model applies to other radioactive isotopes of iodine as well as to other elements with short- and long-lived (or stable) isotopes.

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1. INTRODUCTION

The extents to which the iodine isotopes ^{129}I and ^{131}I are removed from gaseous or liquid streams by various treatment systems are frequently presumed to be different. For example, Yarbro[1] presents retention factors of 20 and 40 for ^{129}I and ^{131}I, respectively, based[2] on the buildup and revaporization of iodine from solutions; such revaporization might reduce the ^{129}I retention relative to ^{131}I by a factor of 2 to 10. Staff members of Allied-General Nuclear Services[3] have presented retention factors in the ratio 5/1 (favoring the retention of ^{131}I); two survey reports[4,5] assign decontamination factors (DFs) of 3000 and 600 for ^{131}I and ^{129}I, respectivwly; and a more recent environmental impact analysis uses DFs of 750 and 75, respectively.[6]

These differences in retention of iodine isotopes are based on the recognition of radioactive decay of ^{131}I (half-life = 8.05 days) as an effective "decontamination" or "retention" factor which, however, does not apply to ^{129}I (half-life = 1.6×10^7 years); some of them are also based on the existence of pathways by which iodine can be recycled within fuel reprocessing plants, thereby effectively reducing decontamination factors. Although recycle and the qualitative difference between short- and long-lived isotopes have been used in analyzing the effectiveness of process equipment, such as silver zeolites, in retaining iodine, apparently no detailed description of the various factors has been given. The purpose of this report is to present such a description in terms of isotopic exchange, material balance equations, and material recycle, and to suggest methods for obtaining the few experimental data needed to ascertain the extent of exchange.

2. THE MODEL AND DEFINITIONS

While the model presented here applies to other elements and their isotopes, definitions are given in terms of a very small mass (a tracer quantity) of a short-lived isotope, ^{131}I, and a relatively very large quantity of nonradioactive isotopes of the same element. In this context, as well as for times less than 1000 years, ^{129}I is treated as "stable."

We consider iodine as one of the components of a gas stream entering a sorption system (see Fig. 1) that can chemically or physically trap a portion of the iodine. Silver zeolite beds and solutions of mercuric nitrate in nitric acid are examples of such systems. Iodine accumulates in the unit, which, eventually, must be regenerated or replaced when its sorption capacity starts to decrease or falls below some specified value. The following definitions apply:

DF = equipment decontamination factor that applies to bulk iodine when the flow pattern is given by Fig. 1(a), Q_1/Q_2;

DF^* = decontamination factor for ^{131}I, q_1/q_2;

DF^s = decontamination factor for stable or long-lived isotope, Q_1/Q_2 [differs from DF when there is recycle, as in Fig. 1(b)];

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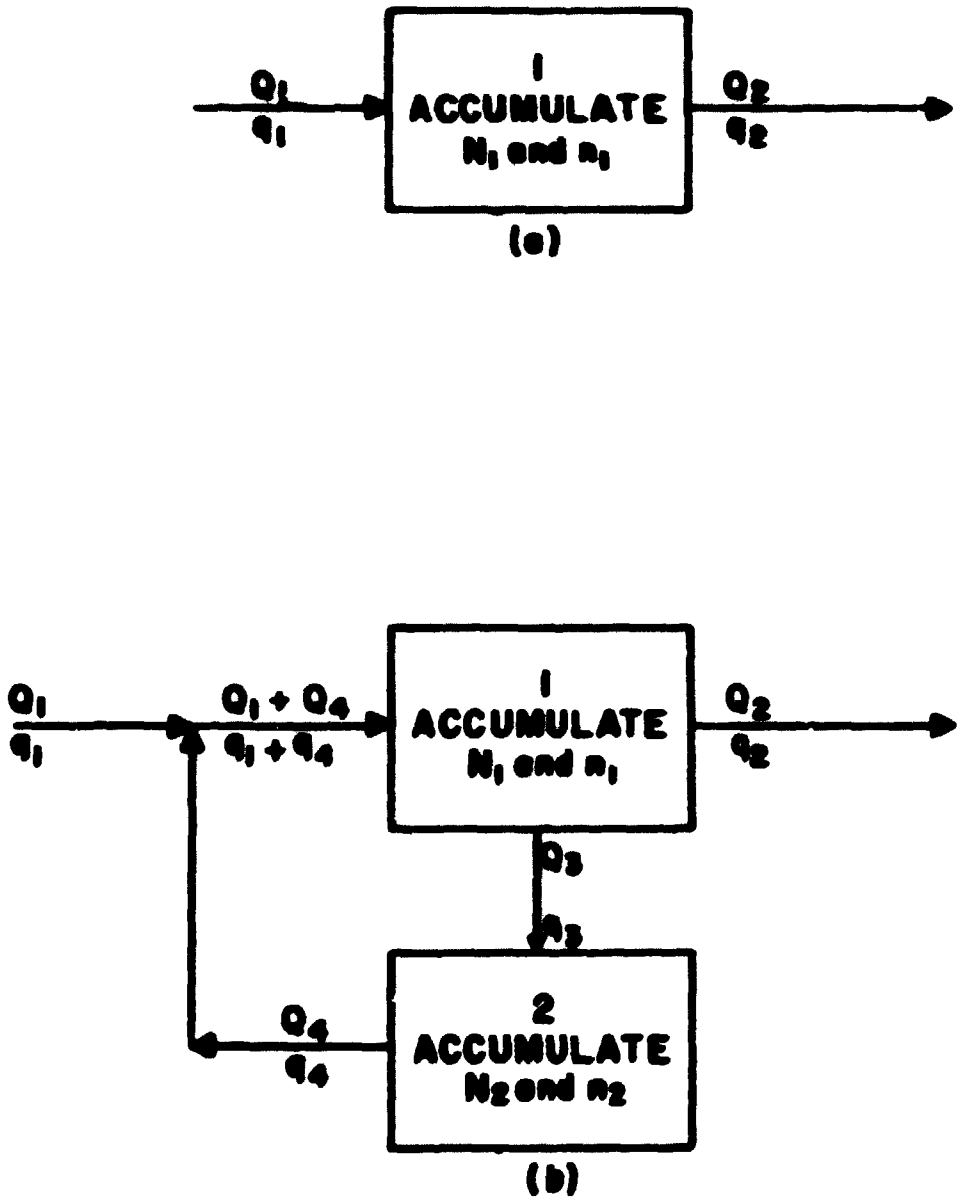

Fig. 1. Flow and accumulation diagrams for sorption units. (a) Without recycle; (b) With one form of recycle.

^{0} = superscript to refer to a value at the zero reference time;

f = fractional attainment of equilibrium isotopic exchange between the gas-phase atoms of iodine in the sorber and those "fixed" on the sorbent before some of the atoms leave the sorber ($0 \leq f \leq 1$);

$k = q_2\,[1 - (1 - f)Q_2/Q_1]$, a combined rate, constant or time-dependent, applicable to Fig. 1(a), day^{-1};

n_1 = number of atoms, or gram-atoms, of ^{131}I "fixed" more or less tightly in the sorbent;

N_1 = number of all atoms, or gram-atoms, of iodine that are "fixed" more or less tightly in the sorbent;

q_1 = the rate, constant or time-dependent, at which ^{131}I enters the sorber, atoms/day or gram-atoms/day (or other time unit);

Q_1 = the rate, considered to be essentially constant (the bulk of the iodine is stable or long-lived ^{129}I), at which all iodine enters the sorber, atoms/day or gram-atoms/day;

q_2 = the rate at which ^{131}I leaves the sorber, atoms/day or gram-atoms/day;

Q_2 = the rate at which all iodine leaves the sorber, atoms/day or gram-atoms/day;

q_3 = the rate at which ^{131}I leaves the sorption unit [Fig. 1(b)] to report to a holdup tank, atoms/day or gram-atoms/day;

Q_3 = the rate, constant or time-dependent, at which iodine leaves the sorption unit 1 [Fig. 1(b)] to report to a holdup tank, atoms/day or gram-atoms/day;

q_5 = the rate at which ^{131}I is recycled to the sorption unit, atoms/day or gram-atoms/day;

Q_5 = the rate at which iodine is recycled to the sorption unit, atoms/day or gram-atoms/day;

t = time, days or any other appropriate unit;

λ = the decay constant of the short-lived species, day^{-1} (or reciprocal of time if another time unit is used) = 0.0861 day^{-1} for ^{131}I;

$t_{1/2}$ = half-life of short-lived isotope, 8.05 days in the case of ^{131}I.

3. NUCLEAR FUEL REPROCESSING

Calculations presented in this report are based on material balance equations and isotopic exchange (which may vary from 0 to 100% in degree of completeness). The material balance equations can be quite numerous, depending on the complexity of the sorption and recycle systems used in a nuclear fuel reprocessing plant to reduce the quantities of iodine, or other element with radioactive isotopes, discharged to the environment. Only two types of sorption systems are analyzed in this report. The first, Fig. 1(a), is a no-recycle system; the second involves recycle in the system shown in Fig. 1(b).

3.1 Sorption Without Recycle

In this case, the material-balance equations for ^{131}I and total iodine entering and leaving a sorbent bed may be written as:

$$dm_1/dt = q_1 - \gamma_2 - \lambda m_1 , \qquad (1)$$

and

$$dM_1/dt = Q_1 - Q_2 - \lambda m_1 . \qquad (2)$$

The term λm_1 in these equations accounts for the loss, by decay, of ^{131}I.

Before extending the mathematical analysis of Eqs. (1) and (2), it is worth noting the magnitudes of the several flow rates. As summarized in Table 1, typical values of the average rates, Q_1, at which all iodine species will report to the primary iodine-removal units in plants reprocessing light-water reactor (LWR) fuels, high-temperature gas-cooled reactor (HTGR) fuels, and liquid-metal-cooled fast breeder reactor (LMFBR) fuels will be about 10 g-atoms/day. The stable isotope ^{127}I and the long-lived ^{129}I (half-life = 1.6×10^7 years) contribute nearly all the mass of Q_1. Similarly, the flow of ^{131}I, q_1, into the primary iodine removal units is in the order of 10^{-7} g-atom/day into LWR and HTGR fuel reprocessing plants, at reprocessing time, and 10^{-4} g-atom/day into the primary sorber of the LMFBR fuel reprocessing plant. If decontamination factors (DFs) for these units are in the order of 10 to 100, the flow,

Table 1. Expected rates of flow of iodine isotopes in LWR, HTGR, and LMFBR fuel reprocessing plants

Flow rate	Type of reactor fuel processed		
	LWR[a]	HTGR[b]	LMFBR[c]
^{131}I, Ci/day	4.6	2.5	3.0 x 10^{3}
q_1, g-atoms/day	2.8 x 10^{-7}	1.5 x 10^{-7}	1.9 x 10^{-4}
^{129}I, g-atoms/day	8.9	8.8	10.3
Total I, Q_1, g-atoms/day	10.5	11.0	13.5

[a]Based on averages from Tables 4.2 and 4.5 of ref. 7 for fuel 160 days after discharge from the reactor. Original output from the ORIGEN code[8] was used as the source of gram-atoms per metric ton of heavy metal (MTHM). The capacity of the LWR fuel reprocessing plant is assumed to be 1500 MTHM/year.

[b]Based on Table 4.8 of ref. 7 for fuel 160 days after discharge from the reactor. Original output from the ORIGEN code[8] was used as the source of gram-atoms/MTHM. The capacity of the HTGR fuel reprocessing plant is assumed to be 450 MTHM/year.

[c]Based on Table 4.14 of ref. 7 for fuel 90 days after discharge from the reactor. Original output from the ORIGEN code[8] was used as the source of gram-atoms/MTHM. The capacity of the LMFBR fuel reprocessing plant is assumed to be 1500 MTHM/year.

q_2, of total iodine out of the primary sorber would be in the range 0.1 to 1 g-atom/day for all of the fuel reprocessing plants; the flows, q_2, of ^{131}I out of the primary sorbers would be in the range 10^{-9} to 10^{-8} g-atom/day in LWR and HTGR fuel reprocessing plants and 10^{-6} to 10^{-5} g-atom/day in the LMFBR fuel reprocessing plant.

The amount, n_1, of ^{131}I retained within a sorbent will be calculated below; however, it is easy to demonstrate that, because of decay, the inventory of this nuclide will not exceed that which flows into the sorbent over a period $t_{1/2}/\ln 2$ (about 11.6 times the daily input rate in the case of ^{131}I). Therefore, n_1 will be less than 3.3 x 10^{-6}, 1.7 x 10^{-6}, and 2.2 x 10^{-2} for LWR, HTGR, and LMFBR fuel reprocessing plants, respectively; the product λn_1 is less than the rate at which ^{131}I enters the plant (Table 1). It is apparent, therefore, that λn_1 will be much smaller than

Q_1 or Q_2 for the primary iodine-removal unit of fuel reprocessing plants. [Exceptions to this occur if DF is $\geq 10^6$ for the LMFBR fuel reprocessing plant or $\geq 10^7$ for LWR or HTGR fuel reprocessing plants; in such instances, $\lambda n_1(max)$ and Q_2 could be nearly equal.] In this case, the term λn_1 could be deleted from Eq. (2) without introducing significant errors, to give:

$$dN_1/dt = Q_1 - Q_2 . \tag{3}$$

Integration of Eq. (3) leads to the very good approximation

$$N_1 = N_1^0 + (Q_1 - Q_2)t . \tag{4}$$

If no exchange occurred between fresh atoms entering the sorber and those already "fixed," the DFs for both ^{131}I and stable isotopes would be equal, and equal to the equipment DF, namely

$$DF^S = DF^* = DF = q_1/q_2 = Q_1/Q_2 ; \tag{5}$$

and

$$q_2 = (q_1/Q_1)Q_2 . \tag{6}$$

If there is complete exchange between atoms in the gas phase and condensed phase in the sorber,

$$q_2 = (n_1/N_1)Q_2 . \tag{7}$$

More generally, for a fractional exchange, f,

$$q_2 = [(1 - f)(q_1/Q_1) + f(n_1/N_1)]Q_2 . \tag{8}$$

On substituting Eq. (8) into Eq. (1), we obtain:

$$dn_1/dt = q_1 - [(1 - f)(q_1/Q_1) + f(n_1/N_1)]Q_2 - \lambda n_1 . \tag{9}$$

Rearrangement of Eq. (9) yields:

$$dn_1/dt + (\lambda + fQ_2/N_1)n_1 = q_1[1 - (1 - f)Q_2/Q_1] . \tag{10}$$

In general, a simple, closed-form solution of Eq. (10) does not exist; however, in several important cases, n_1 may be approximated adequately by a closed-form solution to a simplified form of Eq. (10).

In these cases, a desk calculator can be used to calculate DF^*, the decontamination factor for ^{131}I with a short half-life, as contrasted to values of DF^s, the decontamination factors for stable isotopes (^{127}I) or those with long half-lives (e.g., ^{129}I), which will equal DF.

After an initial shakedown, or startup, period, nuclear fuel reprocessing plants are expected to approach a type of steady state in operations wherein the activity of each radioactive nuclide in spent fuel reporting to head-end operations becomes more or less constant. This condition corresponds to both

$$Q_R = \text{constant} \tag{11a}$$

and

$$Q_R = \text{constant} \ . \tag{11b}$$

Typical values of Q_R for ^{131}I and of Q_R for ^{129}I and the stable ^{127}I for expected operations in LWR, HTGR, and LMFBR fuel reprocessing plants are listed in Table 1.

The system of differential equations described in Eqs. (1) and (2) was integrated numerically using the classical fourth-order Runge-Kutta procedure. Appendix A contains a listing of the FORTRAN-IV program except for the Runge-Kutta subroutine, RKLDEQ, of Nestor.[9] Results of calculations in which only DF was varied and the other parameters were held constant are summarized in Fig. 2. It is apparent that, for any particular DF^s (= DF) for bulk iodine (^{129}I or ^{127}I), the value DF^* for ^{131}I will continue to increase with time as long as complete isotopic exchange occurs; alternatively, if the measured DF^* for ^{131}I is constant, the DF^s for ^{129}I or ^{127}I must be decreasing as long as any isotopic exchange occurs.

The extent of exchange is an important parameter, as shown in Figs. 3 and 4; however, there do not appear to be any data which can be used to test the model of this report and to yield a value for this parameter. The data of Ackley and Davis,[10] discussed below, imply that nearly complete isotopic exchange was achieved in their experiments on trapping of iodine on silver zeolite. "Nearly complete" might mean a value of f as low as 0.8 (perhaps lower) or as large as 1 because of the uncertainties in the experimental values of DF^*.

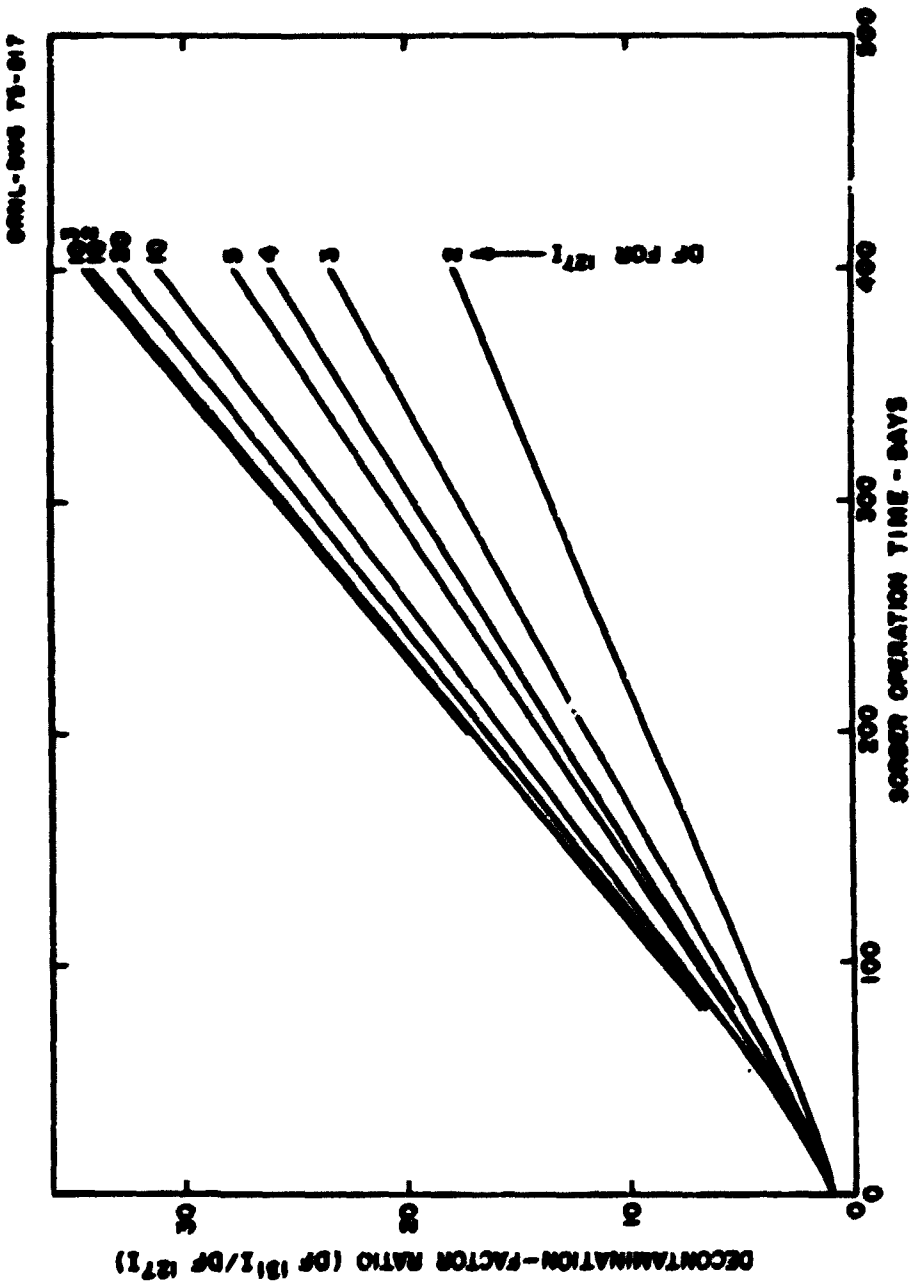

Fig. 2. Effects of time and equipment DF on the ratio DF ^{131}I/DF ^{127}I. Flow pattern is given in Fig. 1(a); f = 1.

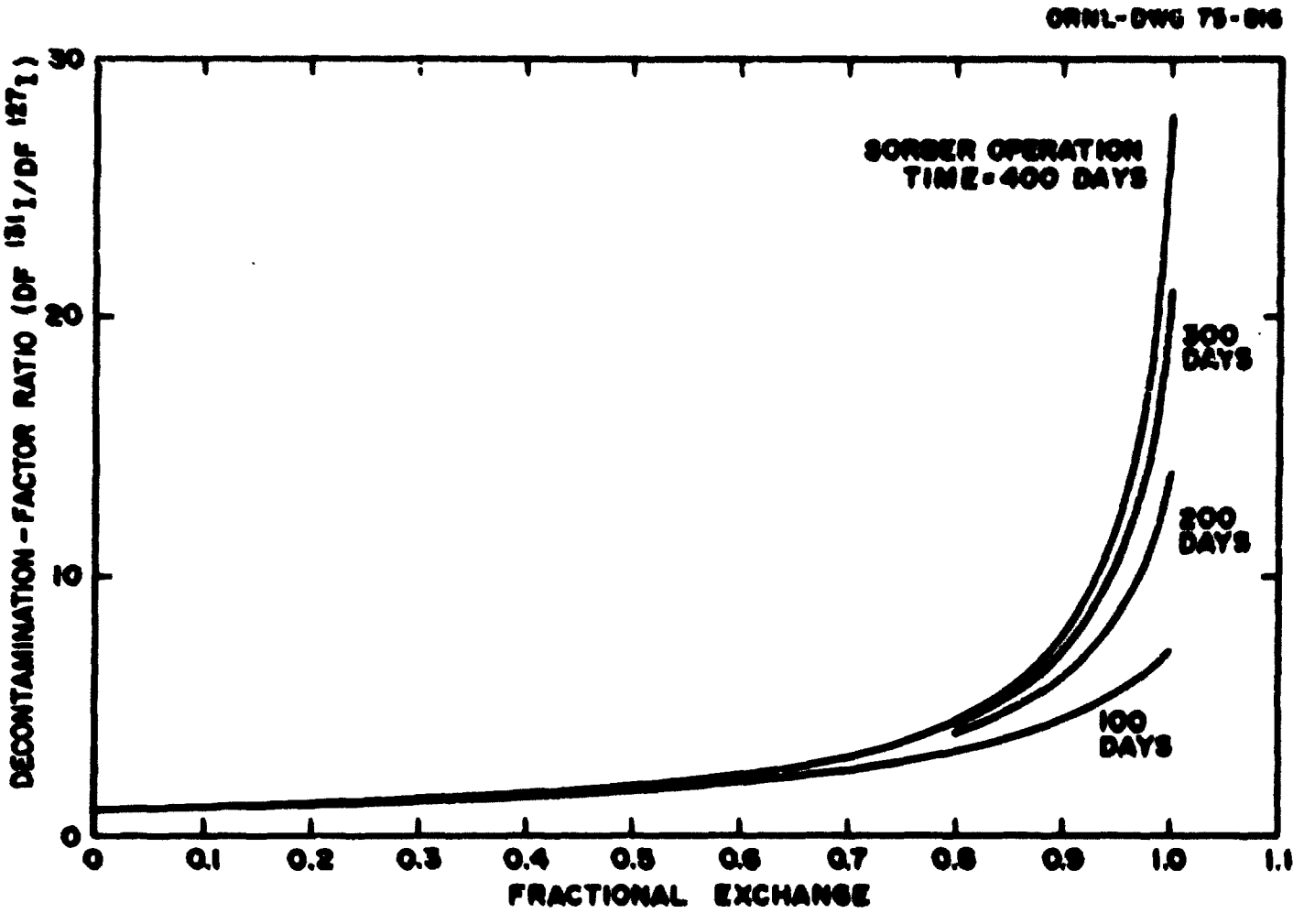

Fig. 3. Effects of fractional exchange and time on the ratio DF ^{131}I/DF ^{127}I. Flow pattern is given in Fig. 1(a); DF (equipment) = 2.

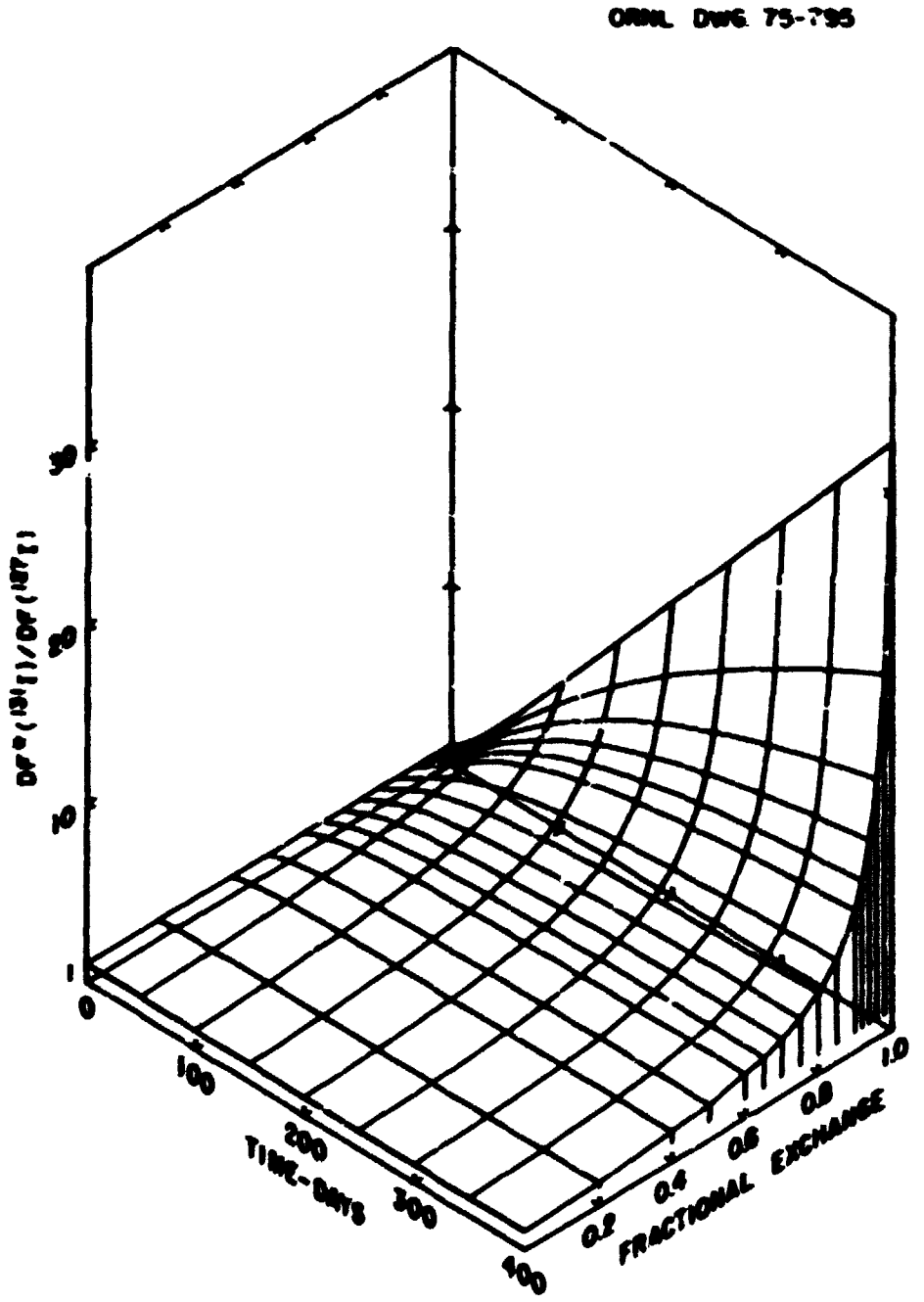

Fig. 4. Effects of time and fractional exchange on the ratio $DF^{131}I/DF^{127}I$. Flow pattern is given in Fig. 1(a).

As mentioned above, Eq. (10) has solutions that may be expressed in closed form for certain boundary conditions. One such situation occurs when $fQ_0/M_1 \ll \lambda$. For ^{131}I, $\lambda = 8.61 \times 10^{-2}$ day^{-1}. Thus, we refer to problems for which $fQ_0/M_1 = fQ_0/[M_1^0 + (Q_1 - Q_0)t] \ll 8.6 \times 10^{-2}$ day^{-1}. When $m_1^0 = 0.$ and $M_1^0 = 0.$, this requires that $(DF - 1)\lambda t \gg f$.

Such a condition is satisfied, for example, if DF^s (stable ^{127}I) exceeds 5 when $f = 1.$ and the sorbent has been in use for 25 days or more; it is also satisfied even for a DF^s value as low as 2 after the bed has been in use for 100 days or more and f is as low as 0.5. For such conditions, Eq. (10) may be replaced with the simpler form

$$dm_1/dt + \lambda m_1 = k \quad . \tag{12}$$

The solution to this equation is:

$$m_1 = \frac{k}{\lambda}(1 - e^{-\lambda t}) \quad . \tag{13}$$

By substituting Eqs. (4) and (13) into Eq. (8), the quantity, q_0, of ^{131}I leaving the sorbent is found to be:

$$q_0 = \left[(1 - f)(q_1/Q_1) + \frac{fQ_0[1 - (1 - f)Q_0/Q_1](1 - e^{-\lambda t})/\lambda}{(Q_1 - Q_0)\,t}\right] Q_0. \tag{14}$$

By using Eq. (5) to define the DF for bulk iodine and using

$$DF^* = q_1/c_0 \ , \tag{15}$$

we obtain

$$DF^* = \frac{1}{\dfrac{(1 - f)}{DF} + \dfrac{f[1 - (1 - f)/DF](1 - e^{-\lambda t})}{(DF - 1)\lambda t}} \quad . \tag{16}$$

Equation (16) summarizes the model of isotopic exchange when $m_1^0 = 0$, $M_1^0 = 0$, and $fQ_0/M_1 \ll \lambda$. Two examples of its use are presented in the subsections that follow.

3.1.1 Values of DF* for the case of no exchange

In this case, f = 0 and DF* = DF at all times.

3.1.2 Values of DF* for the case of complete exchange

In this case, f = 1 and $DF^* = (DF - 1)\lambda t/(1 - e^{-\lambda t})$. Since $(DF - 1)\lambda t \gg 1$ in this example, we find that

$$DF^*/DF \cong 1/DF + (1 - 1/DF)\lambda t \ . \qquad (17)$$

It may be seen from Fig. 2 that the ratio increases as the DF value or time increases; further, the intercepts are nearly 1/DF and the slopes of the linear portions of the curves (t > 100 days) are nearly (1 - 1/DF)λ, as required by Eq. (17). The ratio DF*/DF is nearly independent of DF when DF ≥ 100.

The ratios shown in Fig. 2 for f = 1 increase to about 10 for times in the order of 4 months. Both this factor of 10 and the time period are pertinent in analyzing the performance of process equipment. Expectations or observations of DF*/DF in the order of 10 imply considerable exchange (80% or more) of ^{131}I with the massive quantities of ^{127}I and ^{129}I collected in sorbents; Fig. 2 shows that the factor should increase to approximately 30 after the equipment has performed at design values for a year or more. However, the absence of significantly better performance of a sorbent bed for ^{131}I than for ^{127}I almost certainly corresponds to a small degree (50% or less) of isotopic exchange.

3.2 Sorption With Recycle

The mass-balance equations used in Sect. 3.1 pertain to a specific sorption system [Fig. 1(a)] whose design does not include provisions for recycle of the element of interest, such as iodine. Hence they are not applicable to the many cases of importance in nuclear fuel reprocessing where recycle is an integral step; each of these cases requires individual analysis. Besides the once-through sorption unit of Fig. 1(a), only one other type of sorption system is considered in this report, namely the recycle system of Fig. 1(b). In this system, iodine is sorbed up to

some maximum value, M(max); concurrently, part of the input iodine reports to a reservoir, 2(a waste tank), which may either be the final storage tank or a tank from which iodine can be sparged back to the sorption system. Differential equations needed to describe the system of Fig. 1(b) are more numerous and complicated than those needed to describe the system of Fig. 1(a); however, the calculations can easily be performed when a computer is available.

Using the definitions of Sect. 2, the equations to describe the system of Fig. 1(b) are as follows:

$$dM_1/dt = Q_1 - Q_2 - Q_3 + Q_4 - \lambda m_1 , \tag{18}$$

$$dM_2/dt = Q_2 - Q_4 - \lambda m_2 , \tag{19}$$

$$dm_1/dt = q_1 - q_2 - q_3 + q_4 - \lambda m_1 , \tag{20}$$

$$dm_2/dt = q_2 - q_4 - \lambda m_2 . \tag{21}$$

It is apparent that Eqs. (18) and (20) reduce to Eqs. (2) and (1), respectively, when $Q_3 = Q_4 = 0$.

Equations (18)-(21) can be analyzed only after some boundary conditions have been specified. These conditions may be represented in various ways, such as the flow rates shown for two concepts in Fig. 5. This figure is based on the use of a maximum capacity of the sorbent to trap iodine and on the continuous discharge (as from a mercuric nitrate scrubber discharging to a waste tank) of part of the trapped iodine. The particular example analyzed in this report is based on two assumptions:

(1) The iodine leaves trap 1 and reports to storage tank 2 at a rate given by

$$Q_2 = M_1 U , \tag{22}$$

where the rate constant U is defined as

$$U = \ln 2/T_{y/2} \tag{23}$$

and $T_{y/2}$ is the time required for half the capacity of the sorbent 1 to be expended, perhaps 60 days.

(2) The amount of iodine recycled from unit 2 to the main stream is proportional to the amount of iodine contained in unit 2. In this case,

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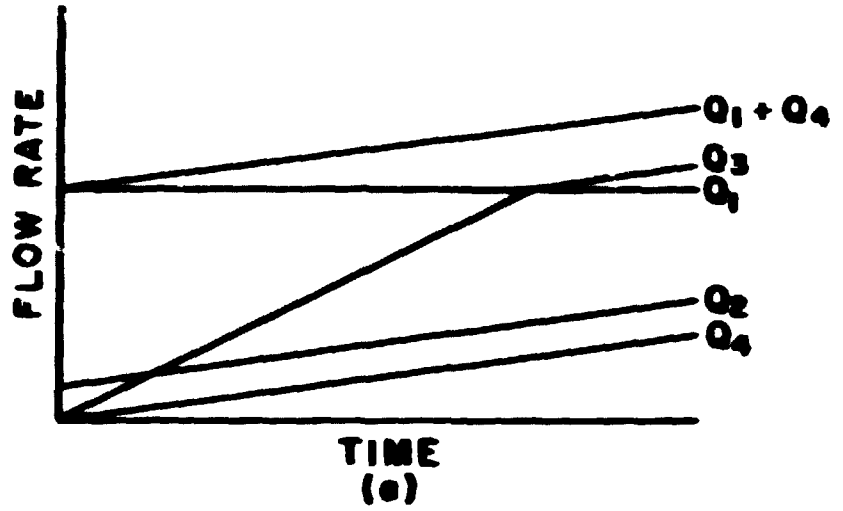


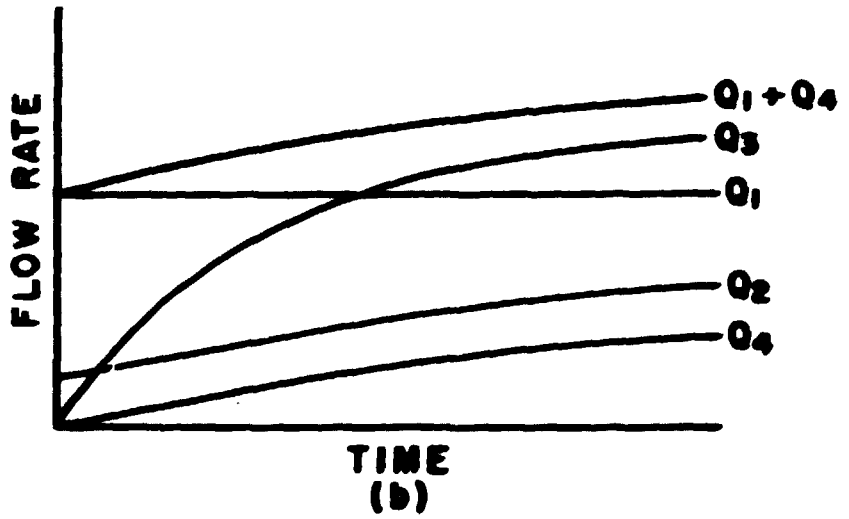


Fig. 5. Two types of flow patterns for the recycle system shown in Fig. 1(b).

$$Q_4 = RM_2 \quad . \tag{24}$$

Since trap 1 is assumed to have a maximum capacity, N(max), the maximum flow of iodine to tank 2 is

$$Q_2(\max) = N_1(\max)\, U \quad . \tag{25}$$

This occurs when trap 1 is saturated with iodine,

$$dM_1/dt = 0 \quad , \tag{26}$$

in which case

$$Q_2 = Q_2(\max) = Q_1 - Q_3 + Q_4 - \lambda n_1 \tag{27}$$

and

$$Q_2(\max) = (Q_1 + Q_4)(1 - 1/DF) - \lambda n_1 \quad . \tag{28}$$

Using Eqs. (22) and (25),

$$Q_2/Q_2(\max) = N_1 U/[N_1(\max)U] \quad . \tag{29}$$

Thus

$$Q_2 = [N_1/N_1(\max)][(Q_1 + Q_4)(1 - 1/DF) - \lambda n_1] \quad . \tag{30}$$

In this same example, we use

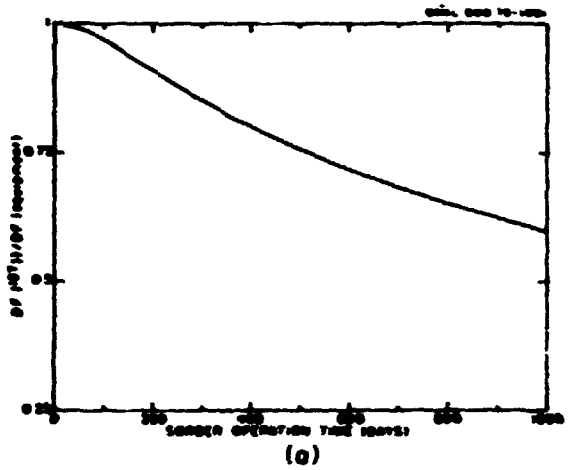
$$q_2 = (n_2/N_2)Q_2 \quad , \tag{31}$$

$$q_4 = (n_4/N_4)Q_4 = Rn_4 \quad , \tag{32}$$

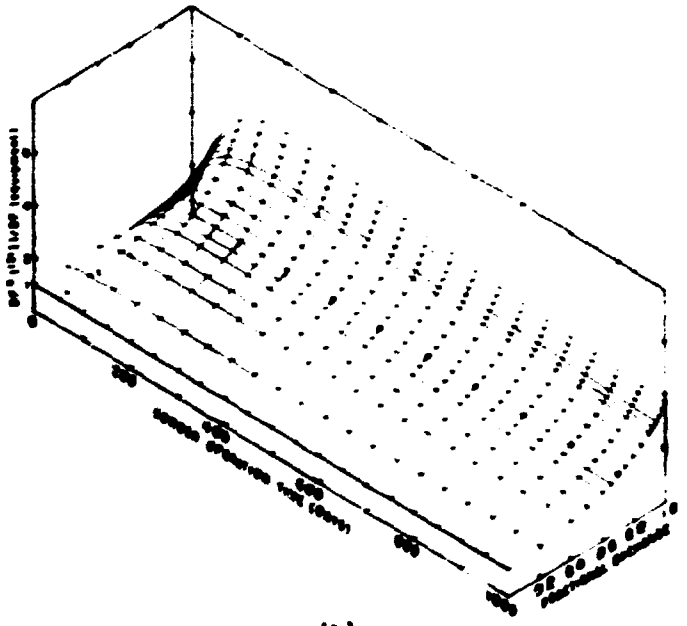
and

$$q_1 = [(1 - f)(q_1 + q_4)/(Q_1 + Q_4) + f(n_1/N_1)]Q_1 \quad . \tag{33}$$

Equations (18)-(33) form the basis of the computer program listed in Appendix A; this program produced the results shown in Figs. 2-4 for a system in which there is no recycle of iodine; it produced the results shown in Fig. 6(a) and 6(b) for a system with recycle.



(a)



(b)

Fig. 6. Effects of time, recycle, and fractional exchange on relative decontamination factors. (a) For ^{127}I; (b) for ^{131}I. In both cases, DF (equipment) = 20, $T_{N(max)}$ = 60 days, R = 1 x 10^{-3}.

4. LABORATORY EXPERIMENTS

Many laboratory studies, such as those of Ackley and Davis,[10] lend themselves to testing the model proposed in this report. In such studies, a quantity of stable iodine is spiked with tracer iodine, such as ^{128}I, ^{130}I, or ^{131}I, which is passed through a sorbent bed until its level of radioactivity becomes too low for accurate counting. In contrast to the situation in Sect. 3, the quantity q_a in these experiments is, therefore, time-dependent according to the relation

$$q_a = q_a^o e^{-\lambda t} \quad ; \tag{34}$$

and Eq. (10) becomes

$$dm_i/dt + (\lambda + fQ_a/M_i)m_i = k^o e^{-\lambda t} \quad , \tag{35}$$

where

$$k^o = q_a^o [1 - (1 - f)Q_a/Q_t] \, . \tag{36}$$

Equation (36) can be integrated analytically to yield

$$m_i = \left\{ \left(m_i^o - \frac{q_a^o M_i}{Q_t} \right) \left[\frac{M_i^o}{M_i} \right]^{fQ_a/(Q_t - Q_a)} + \frac{q_a^o M_i}{Q_t} \right\} e^{-\lambda t} \, . \tag{37}$$

Finally, the quantity DF* is calculated as

$$DF^* = \frac{q_i}{q_a} = \frac{q_a^o e^{-\lambda t}}{q_a} = \frac{DF}{1 + \frac{f}{M_i} \left[\frac{m_i^o Q_t}{q_a^o} - M_i^o \right] [M_i^o/M_i]^{f/(DF-1)}} \, . \tag{38}$$

As examples of the application of the model presented here, either by use of Eqs. (1) and (2), Eq. (10), or Eq. (37), we reanalyze data from Tables 1 and 2 of ref. 10 pertaining to the sorption of iodine by a 26% (Ag^+ for Na^+ exchanged) silver zeolite. The following data are derived from these tables, in the nomenclature of this report. An air stream containing I_2 with tracer ^{126}I($t_{1/2}$ = 60 days) was passed into each of ten test beds for 196 days. At the end of this period, each bed loading was 4.02 mg/ml. The radioactivity level of the tracer was then increased about 500 times and the experiment continued. Samples were taken 90

and 154 days after the ^{128}I activity had been increased, that is, at total elapsed times of 286 and 350 days. Loadings at these times were 7.01 and 9.19 mg/ml, respectively. The flow of I_2 was stopped occasionally during the first 286 days of these experiments, and CH_3I with ^{130}I tracer ($t_{1/2}$ = 12.4 hr) was injected instead. By this method, sorption of both I_2 and CH_3I on the same bed was measured.

In applying Eqs. (37) and (38) to the first 196 days of I_2 sorption, we use the condition that the bed initially contained no iodine. This is equivalent to:

$$n_1^o = N_1^o = 0 \quad . \tag{39}$$

This condition allows Eq. (37) to be simplified to:

$$n_1 = q_2^o (1 - 1/DF)te^{-\lambda t} \quad , \tag{40}$$

and the rate at which ^{128}I was discharged from the bed is given, by use of Eqs. (8), (18), and (40), as

$$q_2 = q_2^o e^{-\lambda t}/DF \quad . \tag{41}$$

The expected result is obtained when q_1 ($= q_2^o e^{-\lambda t}$) is divided by q_2; that is,

$$DF^* = DF$$

for all values of f. Thus, the model does satisfy one expected condition, namely, the decontamination factor obtained for the tracer is the same as that for the bulk iodine, regardless of how complete the exchange might be, as long as a single batch of traced iodine is being used.

When a new batch of traced iodine is used in a system previously exposed to this element, but at a different specific radioactivity of tracer, the decontamination factor for the tracer generally no longer equals the DF for bulk iodine. Instead, $DF^* > DF$ if the specific radioactivity of the new batch is higher than that of the previous one, and $DF^* < DF$ if it is less. The reason for these inequalities may be seen by examining the term $(n_1^o Q_2/q_2^o - N_1^o)$ of Eq. (38). Tracer and bulk

iodine inventories of the sorbent at the end of the usage of one batch of sorbate are n_1^o and M_1^o, respectively. If the bulk iodine flow rate, Q_1, is held constant, an increase in the tracer activity corresponds to an increase in q_1^o, to a decrease in $(n_1^o Q_1/q_1^o - M_1^o)$ and, therefore, to an increase in DF* according to Eq. (38). On the other hand, changing to a sorbate with lower activity corresponds to a decrease in q_1^o, to an increase in $(n_1^o Q_1/q_1^o - M_1^o)$ and, hence, to a decrease in DF*.

Experiments are performed with tracers to utilize particle or photon counting techniques, which are usually much more accurate than normal chemical techniques, especially for obtaining the rate at which a trapped species leaves a highly efficient sorbent bed. Equation (38) cannot be rearranged to solve directly for the bulk-iodine DF for the measured tracer DF*; instead, it is necessary to solve for DF by iterative methods. This can be done with a desk calculator or with the short computer program presented in Appendix B.

Using Eq. (38), a reanalysis of the data of Ackley and Davis[10] pertaining to sorption of iodine by the 26% (Ag^+ for Na^+ exchanged) silver zeolite after the first 196 days of exposure, that is, after changing to iodine with a higher ^{126}I specific activity, shows that DF* increased for constant DF. Some of the calculated values derived from Eq. (38) are listed in Table 2. The DF*/DF ratio, which is in excess of 2.3 after 286 days of exposure and nearly 1.8 at 350 days, is of particular interest. By contrast, Ackley and Davis estimated this ratio to be in the order of 4. Specific activities of sorbed inlet and exit gas-phase iodine, in millicuries per gram of iodine, are also listed in Table 2. Since 100% exchange was assumed, the sorbed-phase specific activity and exit gas-phase activity are equal.

Many of the data in ref. 10 show DF*s for tracer iodine that were too high to measure with the particular specific activity in use. In the example reanalyzed here, the DF*s with the original traced iodine were 1×10^4 and 5×10^3 at 35 and 77 days, respectively; at 140 and 196 days, the DF*s were too large to determine with the low specific activity of ^{126}I in stable iodine. These tracer DF*s are equal to the bulk DFs, as explained above. Data obtained with the higher-activity iodine, introduced

Table 2. Calculations of DF* for stable ^{127}I from DF* values for ^{128}I of Ackley and Davis[10]

Exposure History Through 196 Days			
n_1, inventory of ^{128}I, g-atoms	1.04E-13		
N_1, inventory of ^{126}I + ^{127}I, g-atoms	9.01E-04		
n_1/N_1, inventory ratio	1.16E-10		
Specific activity, mCi ^{128}I/g I	2.00E-03		
Properties of Second Batch of ^{128}I-traced Iodine[a]			
q_2/Q_2, flow-rate ratio	4.15E-08		
Specific activity, mCi ^{128}I/g I	9.58E+00		
Elapsed Exposure Time, days		286	350
Gas-phase Inlet Flow Ratio (q_2/Q_2), g-atoms ^{128}I/g-atom I		2.05E-08	9.73E-09
Gas-phase Inlet Specific Activity, mCi ^{128}I/g I		8.52E-01	1.68E-01
Gas-phase Exit Specific Activity, mCi ^{128}I/g I		1.49E-01	9.42E-02
Sorbed-phase Inventory Ratio (n_1/N_1), g-atoms ^{128}I/g-atoms I		8.65E-09	5.47E-09
Sorbed-phase Specific Activity, mCi ^{128}I/g I		1.49E-01	9.42E-02
Ratio of gas-phase to condensed-phase specific radioactivity [also = $(q_2/Q_2)/(n_1/N_1)$]		2.36	1.78
Reported DF*		4. E+4	9. E+3
Calculated DF (for bulk iodine)		1.69E+4	5.07E+3

[a]The reference time for the second batch is the zero time of the first batch.

at 196 days, were used to calculate additional values of bulk DFs, namely 1.7×10^4 at 286 days and 5.1×10^3 at 350 days. In essence, the data for exposures of 35, 77, 286, and 350 days show bulk DFs of 4×10^3 to 2×10^4; the geometric mean is 8.1×10^3 and the uncertainty is 2.1 to 1/2.1 times the mean. These four values show that, within a factor of 2, the bulk DF for iodine did not change during the 350 days of the test; loading at 350 days was about 9.2 mg of I_2 per milliliter of the 26% (Ag^+ for Na^+ exchanged) silver zeolite. It should be noted that the values for the DF*/DF ratio at 286 and 350 days are based on 100% exchange of iodine nuclides between gas and condensed phases; such complete exchange ($f = 1$) makes the data at the longer times compatible with those at shorter times. Values of f somewhat less than 1 (perhaps as low as 0.9) might similarly be consistent with the data.

The data of Ackley and Davis[10] on CH_3I trapping are not subject to as nearly unique interpretation as are the iodine data. Instead, the following hypotheses are equally valid:

(1) There was no exchange between iodine trapped as I_2 and that introduced as CH_3I. In this case, the tracer DFs (DF*) are equal to the DFs for bulk CH_3I.

(2) Complete exchange occurred between iodine trapped as I_2 and that introduced as CH_3I. In this case, there is no physically significant solution to Eq. (38).

(3) There was partial exchange between iodine trapped as I_2 and that introduced as CH_3I. In this case, as in Case 2, DF* and DF are not equal.

Equation (38) can be greatly simplified when the pertinent experiment involves a very brief injection of one chemical species into a sorbent that already contains sorbate in a different chemical form — CH_3I and I_2 in this case. Since ^{130}I was used to trace CH_3I while ^{128}I was used to trace I_2, the variables in Eq. (38) have the following values: n_1° (^{130}I) = 0 in all CH_3I injection tests; the quantity of iodine deposited as CH_3I was very small compared with that

previously deposited as I_2, in which case $N_1 \simeq N_1^o$, or $N_1^o/N_1 \simeq 1$. With these boundary conditions,

$$\frac{DF^*}{DF} \simeq \frac{1}{1-f} \quad .$$

The ratio of DFs for $CH_3{}^{130}I$ and $CH_3{}^{127}I$ cannot be determined since f is not known. A plot of this ratio vs f is similar to the upper curves in Fig. 3. Thus, for example, DF^*/DF has the values 1., 1.91, and 14.6, respectively, when f assumes the values 0.0, 0.5, and 0.98.

Only in Case 1, which may be valid (but is not known to be), do the data on CH_3I trapping have a simple interpretation. The possibility that Case 3 is valid means, in terms of the present model, that the magnitude of the bulk DF for CH_3I on the 26% (Ag^+ for Na^+ exchanged) silver zeolite cannot be specified. To obtain unambiguous values of this DF for CH_3I, it will be necessary to perform long-term tests, such as those performed with iodine, in which reference data are obtained with CH_3I at two successively higher tracer levels. The extent of exchange of iodine between gaseous and sorbed CH_3I could be determined from such measurements.

5. DISCUSSION AND CONCLUSIONS

The model for trapping of short- and long-lived (including stable) isotopes of a chemical element presented in this report is based on simple concepts, namely, material balances across the trap, material recycle, and isotopic exchange. In the case of iodine trapping, the sorbate could be gaseous iodine in an air stream and the sorbent could be a solution of $Hg(NO_3)_2$ in HNO_3 or a bed of silver zeolite. Material-balance equations such as (1) and (2), or (18)-(21), include loss of a short-lived isotope by decay; the role of exchange is contained in Eq. (8) for a no-recycle system and in Eq. (33) for a particular recycle system. Two applications of the model are discussed in the following paragraphs.

In a nuclear fuel reprocessing plant, the specific activity of ^{131}I will be approximately independent of time after initial shakedown operations. The model predicts, for a no-recycle system, that the decontamination factor

for ^{131}I, namely DF*, will increase uniformly as long as iodine inventory on the sorber is significantly less than the sorber capacity, that is, as long as the decontamination factor for bulk iodine, DF, remains constant and if complete isotopic exchange occurs between iodine in the sorbate and that in the trap. If the exchange is less than complete, the DF*/DF ratio will increase to an approximately constant value that depends on the extent of exchange. Incipient exhaustion of the trap - namely breakthrough - will correspond to a cessation of increase in DF*/DF. Depending on the value of DF for the trap, the DF*/DF ratio could increase to 30 or more during 400 days of operation of a trap for ^{131}I in ^{127}I or ^{129}I; that is, ^{131}I will be removed in the order of 30 times more effectively, because of its radioactive decay, than will the other two isotopes, as shown in Figs. 2 and 4 for complete isotopic exchange. In a process involving recycle, the decontamination factors of all isotopes will be lower than those of a no-recycle system by amounts that can be accurately calculated from the mass-balance equations if the degree of exchange is known.

In a laboratory experiment, the common use over an extended period of time of a batch of traced iodine corresponds to a specific activity that decreases with time. As long as this same batch of traced iodine is used, the decontamination factors for tracer and bulk iodine remain equal, regardless of the degree of chemical exchange. However, when a second batch is introduced into a trap that has been previously exposed to iodine with a lower or a higher specific activity, the decontamination factors for tracer and bulk iodine will not be equal unless there is no isotopic exchange. Laboratory experiments performed sequentially with iodine at two significantly different specific activities can be used to measure both the efficiency of a trap and the degree of isotopic exchange.

The fractional isotopic exchange, f, is a function of physicochemical processes that have not been described in the derivations in this report. Such processes include physical adsorption and chemical reactions on solids, dissolution, and chemical reaction in solutions. All of these processes are temperature-dependent; the chemical reactions are time-dependent. In addition, mass transfer rate could be an important variable

in the evaluation of f. Whereas the number of experiments from which f can be calculated is small, there are apparently no data from which the kinetics of isotopic exchange, on zeolites, in solution scrubbers, etc. can be analyzed. In the absence of such data, it is not possible to define the range over which several process variables, such as flow rates, bed retention time, sorbate concentration, temperature, etc., may change without producing a significant change in f.

6. ACKNOWLEDGMENTS

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7. REFERENCES

1. O. O. Yarbro, Supplementary Testimony Related to Fission Product Source Terms and Effluents from Barnwell Nuclear Fuel Reprocessing Plant, U.S. Atomic Energy Commission, Barnwell Nuclear Fuel Plant, Docket No. 50-332 (October 1974).

2. O. O. Yarbro, Supplementary Testimony Regarding the State of Technology for and Practicability of Control and Retention of Iodine in a Nuclear Fuel Reprocessing Plant, U.S. Atomic Energy Commission, Barnwell Nuclear Fuel Plant, Docket No. 50-332 (October 1974).

3. Allied-General Nuclear Services, Barnwell Nuclear Fuel Plant - Environmental Report, Docket No. 50-332 (November 1971).

4. U.S. Atomic Energy Commission, Fuels and Materials, Directorate of Licensing, Environmental Survey of the Uranium Fuel Cycle, WASH-1248 (April 1974).

5. U.S. Atomic Energy Commission, Fuels and Materials, Directorate of Licensing, Generic Environmental Statement Mixed Oxide Fuel, WASH-1327 (Draft of August 1974).

6. B. C. Finney, R. E. Blanco, R. C. Dahlman, F. G. Kitts, and J. P. Witherspoon, Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing "As Low As Practicable" Guides - Nuclear Fuel Reprocessing, ORNL-TM-4901 (May 1975).

7. J. O. Blomeke, C. W. Kee, and J. P. Nichols, Projections of Radioactive Wastes to be Generated by the U.S. Nuclear Power Industry, ORNL-TM-3965 (February 1974).

8. M. J. Bell, ORIGEN - The ORNL Isotope Generation and Depletion Code, ORNL-4628 (May 1973).

9. W. B. Nestor, Subroutine RKLDEQ for the Integration of First Order Ordinary Differential Equations on the IBM 360/75, ORNL-TM-1762 (March 10, 1967).

10. R. D. Ackley and R. J. Davis, Effect of Extended Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Performance of Sorbents (Final Report), ORNL-TM-4529 (August 1974).

11. W. Davis, Jr., A FORTRAN Program to Draw Isometric-Type Projections with the Calcomp Plotter, ORNL-4127 (August 1967).

12. L. R. Dresner, V. C. Cain, and W. Davis, Jr., INTAKE: A Numerical Program to Calculate Fluid Velocity Profiles near a Rectangular Inlet in a Stream with Cross Flow, ORNL-TM-4185 (July 1973).

8. APPENDIXES

8.1 Appendix A: Fuel Reprocessing Plant Calculations

The computer subroutine FRP listed in this Appendix is based on Fig. 1(b) and Eqs. (18)-(34). Calculations are performed up to statement 151; calculated values are stored for printing or plotting between statements 151 and 401. If the number of cases in a set of data is less than 8, the calculated values are printed according to statements between 401 and 451; if the number of cases in a set of data exceeds 8 (but must not exceed 20 with the listed dimensions), the calculated values of DF'/DF are plotted by use of statements between 451 and 571. The plotting package has previously been described.[11,12] Figures 4 and 6(a) were plotted in this manner.

Input required at statement 41 + 1 is described in comment cards of subroutine FRP; other input at statements 51 and 51 + 1 is associated with terms defined in Sect. 2 and in the following table:

Program variable	Algebraic variable	Program variable	Algebraic variable
DF	DF	Q(1,1)	Q_n
HAFLIF	$t_{1/2}$	QRATIO	q_n/Q_n
EMO(1,1)	M_1^o	TIMEO	Zero time (usually 0.)
EMO(1,2)	m_1^o	RECYCL	R
FRAC	f	TERMAX	Time to load sorbent to N_1 (max)

```
LEVEL  21.6  (DEC '82)                          05/360  PROGRAM  A

          COMPILER OPTIONS - NAME=  MAIN,OPT=02,LINECNT=60,SIZE=0000K,
                                   SOURCE,EBCDIC,NOLIST,NOSOURCE,LOAD,NOMAP,NODECK,NOID,NOXREF
ISN 0002            SUBROUTINE PRP
ISN 0003            IMPLICIT REAL*8(A-H,O-Z)
ISN 0004            DIMENSION AA(5)
ISN 0005            DIMENSION CBASC(20)
ISN 0006            DIMENSION DATA(1200), SPSTAR(250,20), CPSTAR(250,20)
ISN 0007            DIMENSION ES(5,2), SW11(20), SW12(20)
ISN 0008            DIMENSION ESP(5,2), SW21(20), SW22(20)
ISN 0009            DIMENSION ISWE(12)
ISN 0010            DIMENSION LCR(50)
ISN 0011            DIMENSION NSWAP(12)
ISN 0012            DIMENSION C(5,2), Q11(20), Q12(20)
ISN 0013            DIMENSION RATIO1(250,20), RATIO2(250,20), RESVCL(20)
ISN 0014            DIMENSION SWP(20), SPARC(20), SWPLIP(20), ST(250)
ISN 0015            COMMON/PROC/A(5),P(5),C(5),QQ(5),T(5),B,HALF,FE,IWS,F
ISN 0016            COMMON/DESI/T
ISN 0017            COMMON/EE/EES
ISN 0018            REAL*8 AA, B, CQS1, CQS2, D1, D2, D3, DATA, SIG1, SIG2, E0, F0, TT
ISN 0019            REAL*8 E1, E2, TEST
ISN 0020            COMMON/LOCOM/SIG1,SIG2,CQS1,CQS2,D1,D2,D3,A(5),E0(5),F0(5),TT(5)
ISN 0021            COMMON/INPES/NE,NB,NP,NSTCC
                    C     ------------------------------------------
                    C
                    C     ITEB = E      = THE LENGTH OF A TIME SUBSTEP, DAYS FOR ISOTOPE-131
                    C                   = 20 * 0.25  =  5. DAYS AS AN EXAMPLE
                    C
                    C     E             = SINGLE TIME STEP, DAYS FOR ISOTOPE-131
                    C                   = 0.25 DAYS AS AN EXAMPLE
                    C
                    C     NSTL          = NUMBER OF TIME SUBSTEPS
                    C                   = 00 AS AN EXAMPLE
                    C
                    C     NSTL * ITEB * E = TOTAL EXPOSURE TIME
                    C                       = 00 * 20 * 0.25 = 000. DAYS AS AN EXAMPLE
                    C
                    C     P(1)          = DERIV (SW(1,1)) / DT
                    C     P(2)          = DERIV (ES(1,2)) / DT
                    C     P(3)          = DERIV (SW(2,1)) / DT
                    C     P(0)          = DERIV (SW(2,2)) / DT
                    C
                    C     Q(I,J)        = RATE OF FLOW OF COMPONENT J INTO PROCESS UNIT I
                    C     J             = 1  FOR ELEMENTAL TOTAL
                    C     J             = 2  FOR SHORT-LIVED ISOTOPE
                    C
                    C     Y(1)          = ES(1,1)  =  ELEMENTAL TOTAL IN UNIT 1
                    C     Y(2)          = ES(1,2)  =  SHORT-LIVED ISOTOPE IN UNIT 1
                    C     Y(3)          = ES(2,1)  =  ELEMENTAL TOTAL IN UNIT 2
                    C     Y(0)          = ES(2,2)  =  SHORT-LIVED ISOTOPE IN UNIT 2
                    C
                    C     ------------------------------------------
ISN 0022            DWT = 1.000
ISN 0023         61 K = 0
ISN 0024            READ (NT,961) ITEB, NSTL, E
ISN 0025         51 READ (NT,911) DP, HAVLIP, ES0(1,1), ES0(1,2), FRAC, Q(1,1),
                   1                 QRATIO, TIMEO
ISN 0026            READ (NI,911) RECYCL, TEMPAR
ISN 0027            IF (DP - 000) 991, 901, 61
```

```
ISN 0029        61 IF (K .GT. 20) GO TO 901
ISN 0030           TEMP = Q(1,1) * TTEMP1
ISN 0031           PDIF = PRAC - TRAC
ISN 0032           K = K + 1
ISN 0033           TL = ALOG(2.000) / HAFLIF
ISN 0034           PALP = 0.500 * H
ISN 0035           S = 0
ISN 0036           Q(1,2) = QSAVE0 * Q(1,1)
ISN 0037           V(1) = EIG0(1,1)
ISN 0038           V(2) = EIG0(1,2)
ISN 0039           V(3) = 0.
ISN 0040           V(4) = 0.
ISN 0041           T = TIME0
ISN 0042           DO 301 J = 1,NSTL
ISN 0043             DO 151 I = 1,ITER
ISN 0044               Q(0,1) = DECYCL * V(3)
ISN 0045               A(1) = Q(1,1) + Q(0,1)
ISN 0046               Q(0,2) = DECYCL * V(4)
ISN 0047               A(2) = Q(1,2) + Q(0,2)
ISN 0048               A(3) = EL * V(2)
ISN 0049               A(4) = PL * V(4)
ISN 0050               QSAME = A(1) * (DUE - DUE / DF) - A(1)
ISN 0051               Q(2,1) = A(1) / DF
ISN 0052               Q(3,1) = (V(3) / V(1)) * QSAME
ISN 0053               IF (V(1) .GT. 0.0) GO TO 111
ISN 0055               Q(2,2) = (PDIF * A(2) / A(1)) * Q(2,1)
ISN 0056               Q(3,2) = 0.0
ISN 0057               GO TO 101
ISN 0058       111     Q(2,2) = (PDIF * A(2) / A(1) + PRAC * V(3) / V(1)) * Q(2,1)
ISN 0059               Q(3,2) = (V(2) / V(1)) * Q(3,1)
ISN 0060       101     P(1) = Q(1,1) - Q(2,1) - Q(3,1) + Q(0,1) - A(3)
ISN 0061               P(2) = Q(1,2) - Q(2,2) - Q(3,2) + Q(0,2) - A(3)
ISN 0062               P(3) = Q(1,1) - Q(0,1) - A(3)
ISN 0063               P(4) = Q(3,2) - Q(0,2) - A(4)
ISN 0064               CALL RELDEQ
ISN 0065               IF (IERR) 101, 151, 101
ISN 0066       151   CONTINUE
ISN 0067             CAPAC(K) = QMAX
ISN 0068             OPSTAR(J,K) = Q(1,1) / Q(2,1)
ISN 0069             OPSTAR(J,K) = Q(1,2) / Q(2,2)
ISN 0070             EE(1,1) = V(1)
ISN 0071             EE(1,2) = V(2)
ISN 0072             EE(2,1) = V(3)
ISN 0073             EE(2,2) = V(4)
ISN 0074             EE011(K) = EIG0(1,1)
ISN 0075             EE11(K) = EE(1,1)
ISN 0076             EE012(K) = EIG0(1,2)
ISN 0077             EE12(K) = EE(1,2)
ISN 0078             Q11(K) = Q(1,1)
ISN 0079             Q12(K) = Q(1,2)
ISN 0080             RATIO1(J,K) = OPSTAR(J,K) / DF
ISN 0081             RATIO2(J,K) = OPSTAR(J,K) / DF
ISN 0082             PRSTCL(K) = DECYCL
ISN 0083             SDF(K) = DF
ISN 0084             SPRAC(K) = PRAC
ISN 0085             SHF_IP(K) = HAFLIF
ISN 0086             ST(J) = T
ISN 0087       301 CONTINUE
```

```
ISN 0088          GO TO 51
ISN 0089      401 CONTINUE
ISN 0090          IF (K .GT. 0) GO TO 451
ISN 0092          WRITE (NO,1001)
ISN 0093          WRITE (NO,1101) (RHO(I),    I = 1,K)
ISN 0094          WRITE (NO,1111) (RHO11(I),  I = 1,K)
ISN 0095          WRITE (NO,1121) (RHO12(I),  I = 1,K)
ISN 0096          WRITE (NO,1131) (RH1(I),    I = 1,K)
ISN 0097          WRITE (NO,1141) (R12(I),    I = 1,K)
ISN 0098          WRITE (NO,1151) (RFRAC(I),  I = 1,K)
ISN 0099          WRITE (NO,1161) (RHOFLIP(I), I = 1,K)
ISN 0100          WRITE (NO,1171) (RHOOC(I)  , I = 1,K)
ISN 0101          WRITE (NO,1181) (RESVCL(I), I = 1,K)
ISN 0102          WRITE (NO,1201)
ISN 0103          DO 421 J = 2, NUML, 2
ISN 0104            WRITE (NO,1211) ST(J), (RATIO2(J,L),L = 1,K)
ISN 0105      421 CONTINUE
ISN 0106          WRITE (NO,1001)
ISN 0107          WRITE (NO,1221)
ISN 0108          DO 431 J = 2, NUML, 2
ISN 0109            WRITE (NO,1211) ST(J), (RATIO1(J,L),L = 1,K)
ISN 0110      431 CONTINUE
ISN 0111          GO TO 91
ISN 0112      451 READ (NI,911) HEIGHT, WIDTH, ANG1, ANG2, RU12, TICKL, SHADOW
ISN 0113          READ (NI,921) W1, DELX1, W2, DELX2, DIRTY1, DELY1, LTHO
ISN 0114          READ (NI,931) (IRGB(J) , J = 1,12)
ISN 0115          READ (NI,931) (SHRAD(J), J = 1,12)
ISN 0116          READ (NI, 934) NOR, (LOR(L), L = 1,NOR)
ISN 0117          DO 571 LL = 1,NOR
ISN 0118          L = LOR(LL)
ISN 0119          CALL BOX (HEIGHT, WIDTH, ANG1, ANG2, RU12, SHADOW, W1, DELX1,
                 '          W2, DELX2, DIRTY1, DELY1, IRGB, L)
ISN 0120          CALL MAPR (SHRAD, W1, DELX1, W2, DELX2, DIRTY1, DELY1, TICKL)
ISN 0121          DO 521 I = 1,K
ISN 0122            X2 = RFRAC(I)
ISN 0123            CALL CURVE(0.0, X2, 1.0, L, 1)
ISN 0124            DO 511 J = 1,NUML
ISN 0125              X1 = ST(J)
ISN 0126              YPLT = RATIO2(J,I)
ISN 0127              CALL CURVE(X1, X2, YPLT, L, J + 1)
ISN 0128      511     CONTINUE
ISN 0129            CALL CURVE(X1, X2, 1.0, L, J)
ISN 0130      521 CONTINUE
ISN 0131          CALL CURVE(0.0, 0.0, 1.0, L, 1)
ISN 0132          CALL CURVE(ST(NUML), 0.0, 1.0, L, 2)
ISN 0133          CALL CURVE(0.0, 1.0, 1.0, L, 1)
ISN 0134          CALL CURVE(0.0, 0.0, 1.0, L, 2)
ISN 0135          DO 551 I = 10, NUML, 10
ISN 0136            X1 = ST(I)
ISN 0137            DO 541 J = 1,K
ISN 0138              X2 = RFRAC(J)
ISN 0139              YPLT = RATIO2(I,J)
ISN 0140              CALL CURVE(X1, X2, YPLT, L, J)
ISN 0141      541     CONTINUE
ISN 0142            CALL CURVE(X1, 0.0, 1.0, L, K)
ISN 0143      551 CONTINUE
ISN 0144          CALL ADVANC(NI)
ISN 0145      571 CONTINUE
```

```
ISB 0156          GO TO 61
ISB 0157       401 WRITE (60,1001)
ISB 0158       901 FORMAT (2I5,F10.0)
ISB 0159       911 FORMAT (8E10.0)
ISB 0160       921 FORMAT (3(I5,F10.0), I5)
ISB 0161       931 FORMAT (12I2)
ISB 0162      1001 FORMAT (1H1)
ISB 0163      1101 FORMAT (' STABLE-ISOTOPE DP                                ', 1PE10.3)
ISB 0164      1111 FORMAT (' INITIAL STABLE-ISOTOPE INVENTORY,   G-MOLES ', 1PE10.3)
ISB 0165      1121 FORMAT (' INITIAL RADIOACTIVE-ISO. INVENTORY, G-MOLES ', 1PE10.3)
ISB 0166      1131 FORMAT (' ELEMENT TOTAL FLOW RATE,          G-MOLES/DAY ', 1PE10.3)
ISB 0167      1141 FORMAT (' RADIOACTIVE ISOTOPE FLOW RATE,    G-MOLES/DAY ', 1PE10.3)
ISB 0168      1151 FORMAT (' FRACTIONAL ISOTOPIC ENRICHMENT                   ', 1PE10.3)
ISB 0169      1161 FORMAT (' HALF LIFE OF ISOTOPE,                      DAYS ', 1PE10.3)
ISB 0170      1171 FORMAT (' REF CAPACITY FOR ISOTOPE                GRAM-ATOMS ', 1PE10.3)
ISB 0171      1181 FORMAT (' THE FRACTIONAL RECYCLE IN THIS CASE IS           ', 1PE10.3)
ISB 0172      1201 FORMAT ('//' '                                          TIME,
                  1- - -  DP (SHORT HALF-LIFE ISOTOPE) / DP (EQUIPMENT)
                  2 - - ' /   '                                            DAYS
                  3
                  4     ' /)
ISB 0173      1211 FORMAT (' '                                                 1PE10.3)
ISB 0174      1221 FORMAT ('//' '                                          TIME,
                  1-   DP (LONG HALF-LIFE OR STABLE ISOTOPE) / DP (EQUIPMENT)
                  2 - - ' /   '                                            DAYS
                  3
                  4     ' /)
ISB 0175          RETURN
ISB 0176          END
```

7. Appendix B: Calculations Based on Laboratory Data

The computer subroutine LABEV listed on the next three pages was written to solve Eqs. 41 and 42 ; calls for input between statements -1 and 91 were chosen to utilize the data of Ackley and Davis.[10] Figure 1(a) is a model of the flow pattern; algebraic and program variables are related in the following table:

Program variable	Algebraic variable	Program variable	Algebraic variable
TIME	Elapsed time when a sample was taken for counting, t	DFSEXP	DF* (experimental)
FLORAT	Gas flow rate for a single sorbent unit	HALLIF	$t_{1/2}$
CONC	Conc. of I_2 in the inlet air	ENO1	N_1^o
QRATIO	Q_2/Q_1	ENO2	n_1^o
DATWT	Atomic weight of tracer	FRAC	f
DATATS	Atomic weight of bulk iodine	TIMEO	Time at start of exposure. In a run of successive samples, TIMEO is set to -1 after the first sample; this causes the program to set TIMEO to the previous sample time.

```
LEVEL  21.6  (DEC 72)                    OS/360  PORTRAN H

          COMPILER OPTIONS - NAME= MAIN,OPT=02,LINECNT=60,SIZE=00064,
                             SOURCE,EBCDIC,NOLIST,NODECK,LOAD,MAP,NOEDIT,REAL,NOREF
   ISN 0002           SUBROUTINE LABEL
   ISN 0003           IMPLICIT REAL*8(A-H,O-Z)
   ISN 0004           DIMENSION ACTESE(25,10), ACTGSO(25,10), ACTSEO(25,10)
   ISN 0005           DIMENSION DPSCAL(25,10), RATIO(25,10)
   ISN 0006           DIMENSION SDP(25,10), SEW01(25,10), SEW02(25,10)
   ISN 0007           DIMENSION SEW1(25,10), SEW2(25,10), SFRAC(25,10), SNAPLIP(10)
   ISN 0008           DIMENSION SQ11(25,10), SQ012(25,10), SQ12(25,10), ST(25)
   ISN 0009           COMMON/TAPES/NI,NO,NP,NTTOC
   ISN 0010           WRITE (NO,1001)
   ISN 0011           CON1 = 6.021 D 23 * DLOG(2.000) / 3.7 D 10 / 86400.
   ISN 0012           CONVRG = 1.00-6
   ISN 0013           ONE = 1.000
   ISN 0014           ICOUNT = 0
   ISN 0015           J = 0
   ISN 0016        61 J = J + 1
   ISN 0017           READ (NI,901) TIME, FLORAT, CONC, QRATIO
   ISN 0018           READ (NI,901) GATWT, GATWTS
   ISN 0019           K = 0
   ISN 0020        51 READ (NI,901) DPSEED, SNAPLIP, EW01, EW02, FRAC, TIME0
   ISN 0021           DP = DPSEED
   ISN 0022           IF (DP - ONE) 601, 401, 61
   ISN 0023        61 IF (K .GT. 6) GO TO 401
   ISN 0025           K = K + 1
   ISN 0026           EL = DLOG(2.000) / SNAPLIP
   ISN 0027           Q11 = FLORAT * CONC * 1000.D0
   ISN 0028           Q012 = QRATIO * Q11
   ISN 0029           Q11 = Q11 / GATWT
   ISN 0030           Q012 = Q012 / GATWTS
   ISN 0031           IF (TIME0 .LT. 0.) Q012 = Q012 * DEXP(-EL*ST(J-1))
   ISN 0033           IF (TIME0 .LT. 0.) TIME0 = ST(J-1)
   ISN 0035           IF (EW01 .LT. 0.) EW01 = SEW1(J-1,K)
   ISN 0037           IF (EW02 .LT. 0.) EW02 = SEW2(J-1,K)
   ISN 0039           DTIME = TIME - TIME0
   ISN 0040           Q21 = Q11 / DP
   ISN 0041           DIF = Q11 - Q21
   ISN 0042           EW1 = EW01 + DIF * DTIME
   ISN 0043           ICK = 0
   ISN 0044        81 ICK = ICK + 1
   ISN 0045           IF (ICK .GT. 10) GO TO 121
   ISN 0047           EXP1 = FRAC / (DP - ONE)
   ISN 0048           TERM1 = (EW01 / EW1) ** EXP1
   ISN 0049           TERM2 = EW02 * Q11 - Q012 * EW01
   ISN 0050           TERM3 = TERM2 / Q11
   ISN 0051           EXPT = DEXP(-EL * DTIME)
   ISN 0052           EW2 = (TERM1 * TERM3) + Q012 * EW1 / Q11) * EXPT
   ISN 0053           TERM = FRAC * TERM2 * TERM1 / EW1
   ISN 0054           DPSCAL(J,K) = DP * Q012 / (Q012 + TERM)
   ISN 0055           T1 = DPSEED / DPSCAL(J,K)
   ISN 0056           T2 = T1 - ONE
   ISN 0057           DP = DP * T1
   ISN 0058           IF (DABS(T2) - CONVRG) 131, 131, 81
   ISN 0059       121 WRITE (NO,1301) J, K, T1, DPSEED
   ISN 0060       131 Q21 = Q11 / DP
   ISN 0061           DIF = Q11 - Q21
   ISN 0062           EW1 = EW01 + DIF * DTIME
   ISN 0063       151 CONTINUE
```

```
ISN 0064            Q12 = Q012 * EXPT
ISN 0065            Q22 = Q12 / DPSCAL(J,I)
ISN 0066          251 RATIO(J,I) = DPSCAL(J,I) / DP
ISN 0067            ST(J) = TIME
ISN 0068          301 CONTINUE
ISN 0069            SER01(J,I) = ER01
ISN 0070            SER1(J,I) = ER1
ISN 0071            SDP(J,I) = DP
ISN 0072            SPRAC(J,I) = PRAC
ISN 0073            SAMPLIP(I) = AMPLIP
ISN 0074            SER02(J,I) = ER02
ISN 0075            SER2(J,I) = ER2
ISN 0076            ACTDOB = ER2 * CONV / AMPLIP
ISN 0077            ACTDOB(J,I) = ACTDOB / (ER1 * GATWT)
ISN 0078            ACTGSI(J,I) = Q12 * CONV / AMPLIP
ISN 0079            ACTGSI(J,I) = ACTGSI(J,I) / (Q11 * GATWT)
ISN 0080            ACTGSO(J,I) = Q22 * CONV / AMPLIP
ISN 0081            ACTGSO(J,I) = ACTGSO(J,I) / (Q21 * GATWT)
ISN 0082            SQ11(J,I) = Q11
ISN 0083            SQ12(J,I) = Q12
ISN 0084            SQ012(J,I) = Q012
ISN 0085            GO TO 51
ISN 0086          401 CONTINUE
ISN 0087            ICOUNT = ICOUNT + 1
ISN 0088            IF (ICOUNT .LE. 3) GO TO 411
ISN 0090            ICOUNT = 1
ISN 0091          411 CONTINUE
ISN 0092            WRITE (NO,1101) (SDP(J,I),   I = 1,K)
ISN 0093            WRITE (NO,1111) (SER01(J,I), I = 1,K)
ISN 0094            WRITE (NO,1121) (SER1(J,I),  I = 1,K)
ISN 0095            WRITE (NO,1131) (SER02(J,I), I = 1,K)
ISN 0096            WRITE (NO,1141) (SER2(J,I),  I = 1,K)
ISN 0097            WRITE (NO,1151) (SQ12(J,I),  I = 1,K)
ISN 0098            WRITE (NO,1159) (SQ11(J,I),  I = 1,K)
ISN 0099            WRITE (NO,1171) (SPRAC(J,I), I = 1,K)
ISN 0100            WRITE (NO,1181) (SAMPLIP(I), I = 1,K)
ISN 0101            WRITE (NO,1201) ST(J),(DPSCAL(J,I),  I = 1,K)
ISN 0102            WRITE (NO,1211) (RATIO(J,I), I = 1,K)
ISN 0103            WRITE (NO,1221) (ACTSDB(J,I),I = 1,K)
ISN 0104            WRITE (NO,1231) (ACTGSI(J,I),I = 1,K)
ISN 0105            WRITE (NO,1241) (ACTGSO(J,I),I = 1,K)
ISN 0106            WRITE (NO,1291)
ISN 0107          501 CONTINUE
ISN 0108            IF (ICOUNT .EQ. 3) WRITE (NO,1001)
ISN 0110            GO TO 61
ISN 0111          601 WRITE (NO,1001)
ISN 0112          901 FORMAT (8E10.0)
ISN 0113         1001 FORMAT (1H1)
ISN 0114         1101 FORMAT (' STABLE-ISOTOPE DP                              ',
                     1  10X, 1P6E10.3)
ISN 0115         1111 FORMAT (' INITIAL ELEMENT INVENTORY,          G-MOLES ',
                     1  10X, 1P6E10.3)
ISN 0116         1121 FORMAT (' FINAL ELEMENT INVENTORY,            G-MOLES ',
                     1  10X, 1P6E10.3)
ISN 0117         1131 FORMAT (' INITIAL RADIOACTIVE-ISO. INVENTORY, G-MOLES ',
                     1  10X, 1P6E10.3)
ISN 0118         1141 FORMAT (' FINAL RADIOACTIVE-ISO. INVENTORY,   G-MOLES ',
                     1  10X, 1P6E10.3)
```

```
ISN 0119      1151 FORMAT (' EFFLUENT TOTAL FLOW RATE,          G-MOLES/DAY ',
                  1  10X, 1P6E10.3)
ISN 0120      1161 FORMAT (' RADIOACTIVE ISOTOPE FLOW RATE,  G-MOLES/DAY ',
                  1  10X, 1P6E10.3)
ISN 0121      1171 FORMAT (' FRACTIONAL ISOTOPIC ENRICHMENT,                ',
                  1  10X, 1P6E10.3)
ISN 0122      1181 FORMAT (' HALF LIFE OF ISOTOPE                     DAYS ',
                  1  10X, 1P6E10.3)
ISN 0123      1201 FORMAT (' SAMPLING TIME AND CALC. DP*,   DAYS AND DP* ',
                  1        1P7E10.3)
ISN 0124      1211 FORMAT (' DECONTAMINATION FACTOR RATIO,        DF*/DF ',
                  1  10X, 1P6E10.3)
ISN 0125      1221 FORMAT (' SPEC. ACTIV. OF SORBATE ON SORBENT,    CI/G ',
                  1  10X, 1P6E10.3)
ISN 0126      1231 FORMAT (' SPEC. ACTIV. OF SORBATE, INLET STREAM, CI/G ',
                  1  10X, 1P6E10.3)
ISN 0127      1241 FORMAT (' SPEC. ACTIV. OF SORBATE,OUTLET STREAM, CI/G ',
                  1  10X, 1P6E10.3)
ISN 0128      1291 FORMAT (//)
ISN 0129      1301 FORMAT (' CONVERGENCE OF CALCULATED DF* TO EXPERIMENTAL VALUE HAS
                  1NOT ACHIEVED IN 30 ITERATIONS. ' /
                  2        ' THE LAST VALUE OF (DFSEXP/DFSCAL(', I2, I2, ') WAS ',
                  3        :P2E10.3, ' FOR DFSEXP = ', E10.3)
ISN 0130            RETURN
ISN 0131            END
```